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IMPLICATIONS FOR THE CRYOGENIC FIELDING
OF LEAKING BERYLLIUM CAPSULES

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In this paper we show that the ambient temperature measured leakage time constant, τ_{RT}, is related to the leakage at cryogenic temperature, R_C, by

$$R_C = 0.23 \rho_{DT} V_{sh} / \tau_{RT}$$

where ρ_{DT} is the density of cryogenic DT vapor, and V_{sh} is the internal volume of the shell. We then calculate the size of voids that may result from leakage at the Be/DT interface, depending upon the number of leakage sites and τ_{RT}. Even for the slowest leakers the potential void growth is excessive. Reasons that voids have not been seen in DT layering experiments to date include the lack of a technique to see isolated micronish bubbles, however possible mechanisms preventing void formation are also discussed.

I. INTRODUCTION

The current point design capsule to be used at the National Ignition Facility[1] in the ignition campaign to begin in 2009 is a beryllium (Be) capsule with graded copper (Cu) doping.[2] These capsules are produced by sputtering Be and as needed Cu onto spherical shell mandrels agitated in a pan below the sputtering guns.[3] Nearly all of the very rigid dimensional and compositional specifications have been met.[4] However very recently it was discovered that the "standard" procedure for fabricating these capsules produced ones that were not gas tight.[5] The room temperature leakage time constant, τ, is defined by

$$p(t) = p_0 e^{-t/\tau} \tag{1}$$

where $p(t)$ is the time dependent internal pressure of a capsule filled at time $t = 0$ to a pressure p_0. It was found that τ was typically only a few minutes to hours. However, two 0.25 μm layers of 10-15% boron doped Be in the first micron of coating largely solved this problem, resulting in many shells with τ's of a week or more, presumably by forming an amorphous permeation barrier.[5] However, even with this dramatic improvement, there is still scatter in the data, shells from the same coating run often have very different measured leakage time constants. From a practical point of view, all shells that will be used as targets in the ignition campaign will be leak-tested. However the current testing technique can only detect leakage from shells with time constants of about 60 days or less.[5,6]

This paper explores the possible implications for a slowly leaking shell with a room temperature half-life of up to 1 year. We first, however, will show the relationship between the room temperature measured τ and cryogenic temperature leak rates. Following this we will explore what the effects might be if the leakage leads to voids in the solid DT at the inner surface of the shell. Lastly we will discuss possible scenarios that may ameliorate the leakage problem.

II. LEAKAGE AT CRYOGENIC TEMPERATURE

The leakage measurements are made at room temperature with deuterium (D_2) gas. By either directly following the time dependence of the leaking D_2 with a mass spectrometer, or for a slow leaker comparing the mass spectrometer signal with a known standard, the time constant τ, as defined in Eq. (1) is determined.[6] In this case the gas pressure in the shell decreases with time, and thus the leak *rate* also decreases.

It will be assumed that the leakage paths are the same at cryogenic temperature (~20 K) as they are at ambient temperature (~300 K). To first order this is probably true; there will be a small amount of contraction as the shell is

cooled, however there is no reason to assume that the leakage pathways will close up. This assumption is supported by leak rate studies done at both liquid nitrogen (77 K) and ambient temperature on the same shell.[5] It was found that the ratio of the measured time constants was about 2,

$$\frac{\tau_{77\,K}}{\tau_{300\,K}} \cong 2 \cong \sqrt{\frac{300}{77}} , \qquad (2)$$

indicating molecular flow (molecular velocity $\propto \sqrt{T}$) and unchanging leakage pathways. The finding of molecular flow, as opposed to viscous flow where there is essentially no temperature dependence,[5] is important to the treatment below.

Further we will assume that the leakage is of DT gas subliming from solid DT. Initially the leakage pathways are filled with solid DT. However the DT will sublime from the ends of these paths, and for this exercise we will take the initial time to be the point at which the pathway is free of solid DT. Since the leaking DT vapor is in equilibrium with solid DT, the vapor pressure doesn't change as the leakage proceeds. Thus our first task is to relate the room temperature measured time constants to the actual leakage *rate* at cryogenic temperature.

We can write Eq. (1) in terms of moles, n, as

$$n = n_0 e^{-t/\tau_{RT}} \qquad (3)$$

where I have added the subscript RT (Room Temperature) to remind us that the measured time constant is at ambient temperature. Equation (3) can be differentiated with respect to time and evaluated at $t = 0$ to give the initial room temperature rate, R_{RT} as

$$-\left.\frac{dn}{dt}\right|_{t=0} = R_{RT}(t=0) = \frac{n_0}{\tau_{RT}} = \frac{(n_0/V_{sh}) \cdot V_{sh}}{\tau_{RT}} \qquad (4)$$

where V_{sh} is the volume of the shell. Note here that n_0/V_{sh} is simply the gas density. At cryogenic temperature the DT vapor density is determined by the temperature of the solid and is thus a *constant* as the leakage occurs.

We can write the leakage rate at cryogenic temperature, R_C, in terms of the leak rate at room temperature as

$$R_C = \sqrt{\frac{T_C}{T_{RT}}} \cdot \sqrt{\frac{M_{D_2}}{M_{DT}}} \cdot R_{RT} . \qquad (5)$$

Thus

$$R_C = \sqrt{\frac{20}{300}} \sqrt{\frac{4}{5}} \cdot R_{RT} = 0.23 \frac{\rho_{DT}(T) \cdot V_{sh}}{\tau_{RT}} \qquad (6)$$

where the first term under the root sign in Eq. (5) is the temperature effect of slowing the molecular velocity, and the second term is the mass effect (DT vs. D_2) which also slows the average velocity. Note that ρ_{DT} is the *vapor* density. Thus from Eq. (6) we can compute the rate of DT loss from a cryogenic shell at a given cryogenic temperature from the room temperature measured time constant.

Let's look first at what this might mean for the current specification of $\tau_{RT} = 10$ days.[7] Here, and in all subsequent calculations, we will take the relevant cryogenic temperature as 0.5 K below the triple point, or 19.29 K, where the vapor density is 0.5 mg/cm^3. The volume of the shell is 2.48 x 10^{-3} cm^3,[8] thus the leakage rate, R_C, is 2.9 x 10^{-5} mg/day. Since the DT layer is about 75 μm thick, and the density of the solid is 250 mg/cm^3, this leakage rate represents only a 0.02% loss per day. This is certainly not a problem, especially since other requirements indicate the shot needs to take place within 30 h of filling to avoid an excessive build up of He.[2]

It is also informative to calculate the limits of the current He leak detection apparatus that is used in cryogenic experiments. At 20 K the detection limit is about 10^{-10} mbar-liter/s,[9] which is 6 x 10^{-14} mol/sec. The He pressure pumped into the shells is 100-200 Torr, similar to the vapor pressure of DT at 19.29 K. If the leakage rate of He is similar to DT, then the maximum τ_{RT} it could detect is about 10 days. Thus slower leakers would appear as non-leakers.

III. POSSIBLE EFFECTS OF LEAKAGE ON THE Be – DT INTERFACE

Let us now look more closely at what may happen when the DT leaks out of small orifices, specifically what is the result at the DT/Be wall interface. Let us assume that as the DT leaks out, a small void forms in the solid DT at the wall, its volume equal to the amount of DT which has left. We will consider two cases, one in which there are only 10 leakage sites, and the other in which there are 10000 sites. In this latter case the leakage sites are on average 30 μm apart, still large compared to the

approximately 1 μm lateral grain dimension. We will also consider two room temperature leakage time constants, $\tau_{RT} = 10$ days, and $\tau_{RT} = 500$ days, this latter representing a half-life of almost 1 year, and much longer than can be currently measured.[6] We will assume that the leakage rate is the same from each orifice *i.e.* $R_C/10$ or $R_C/10000$ per site for shells with 10 or 10000 leakage sites. Clearly the volume of individual voids in the latter case will be 1/1000 of the volume of individual voids in the former.

What will be the size of these voids? Let's look at the 10 site situation with $\tau_{RT} = 10$ days. The rate of loss at each site is 2.9 x 10^{-6} mg/day which represents a solid volume of 1.14 x 10^4 μm^3 per day. This is huge! It would be a sphere of diameter 28 μm. If there were 10000 sites the void volume would be reduced by a factor of 1000, but the diameter by only a factor of 10 giving a 2.8 μm diameter. Even with $\tau_{RT} = 500$ days the respective values are 7.6 and 0.76 μm for the diameter of spherical voids after 1 day. Though the growth in volume is linear with time, the growth in diameter goes as (time)$^{1/3}$. The situation is plotted in Fig. 1 for the four cases discussed.
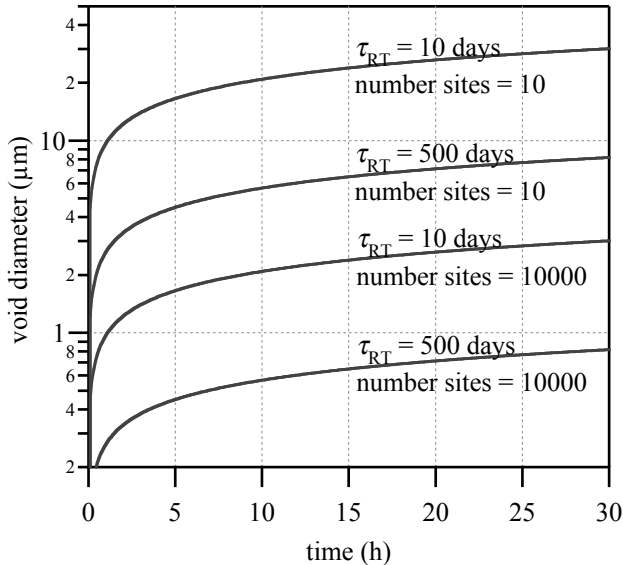

Figure 1. Shown is the computed diameter growth in μm of individual spherical voids at the Be/DT interface due to leakage. The four cases shown are for $\tau_{RT} = 10$ and 500 days and for 10 and 10000 leakage sites.

At present there is no "isolated" defect specification for the inner shell surface, but probably a 10 μm void would not be acceptable. There is, however, a roughness specification,[2] and we can look at the effect of 10000 growing void defects on the inner surface. Shown in Fig. 2 is the evolution of the rms roughness in nm for the case with 10000 sites and either $\tau_{RT} = 10$ or 500 days. The rms roughness specification for modes 100 and higher, which encompasses the roughness that these voids potentially produce, is about 5 nm.[2]
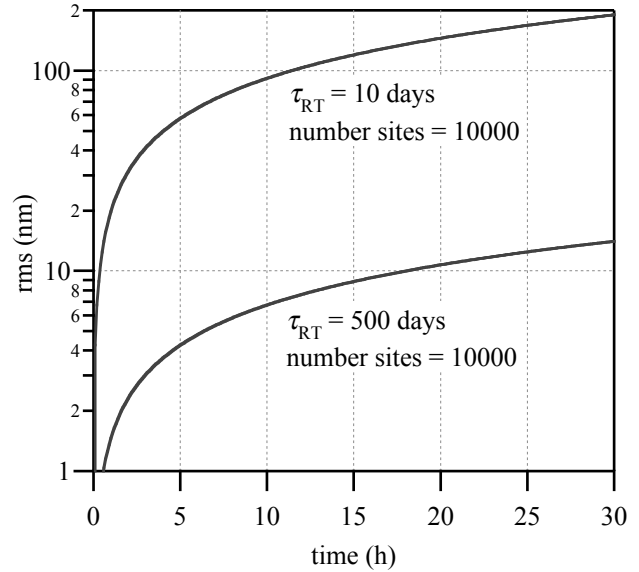

Figure 2. Shown is the time dependent roughness that would be incurred if 10000 spherical voids grew at the Be/DT interface, for shells with room temperature leakage time constants of 10 and 500 days. The inner surface roughness specification over modes greater than 100 is about 5 nm rms.[2]

What is clear is that if void defects form at the inner Be/DT interface, even with shells that have year long half-lives, the evolution on inner surface roughness is a concern.

IV. DISCUSSION

Voids have not been seen at the inner surface in cryogenic layering experiments to date.[9] Why not? There are several possible reasons. First it is possible that the Be shells used to date, and there have been relatively few, are non-leakers. Certainly rapid leakers have not been used. Second, perhaps there is physics that makes a shell which is a slow leaker at room temperature, a non-leaker at cryogenic temperature. For example there may be interactions in the narrow pathways that prohibit or at least delay leakage from the inner fuel. Third, and perhaps more plausible, the shells used have been slow leakers, and voids at the interface have not been seen because the detection system, phase contrast x-ray radiography, has a resolution limit of about 10 μm.[9] It is unlikely that a void at the interface would grow to detectable size before it "detached" and traveled radially toward the inner solid DT surface. The rate of detach-

ment and the size of the detached DT vapor bubbles would depend on their rate of growth. One might imagine a stream of DT vapor bubbles heading toward the inner solid surface. For example, for a $\tau_{RT} = 500$ days and 10 leakage sites, a 1 μm diameter vapor bubble would form every 3.3 minutes.

There are two other reasons, related to the DT itself. First, it is possible that the elastic constants of the DT are such that voids are continually "removed" by flows of material, driven by surface tension effects. Solid DT is quite "soft" particularly near the triple point, and possibly voids never form, the "solid" simply fills the space vacated by sublimed molecules. The second reason, possibly related to this, is that the self-diffusion constant of solid DT is very high. At 0.5 K below the triple point the value of the self diffusion constant, D, is[10]

$$D = \frac{r^2}{t} = 516 \ \mu m^2/h \qquad (7)$$

which means that in an hour the average distance traveled by a DT molecule is about 23 μm. This is in part due to a very high fraction of vacancies in the solid, on the order of 8 x 10^{-4} at 19.29 K.[11] How does this compare to the leak rate – might molecular voids simply diffuse away? The total number of molecules of DT in a 75 μm thick layer is about 1.8 x 10^{19}, thus there are about 1.5 x 10^{16} vacancies in the solid. With $\tau_{RT} = 500$ days, there is a loss at cryogenic temperature of 6.8 x 10^{13} molecules per day, only 0.5% of this number. Of course the molecular voids are all created at the site of the leakage, thus a non-equilibrium number would build up around leakage sites, and depending upon void diffusion versus nucleation rates bubbles may form. This would be more likely if there were only a few such sites, and of course more likely as τ_{RT} decreases.

V. CONCLUSION

We have shown the relationship between the room temperature measured leak rate of Be shells and the respective cryogenic leak rate. The effects of possible leakage of DT in the Be shells is not a problem in terms of loading and routine fielding if the leakage half-life is at least 7 days. There are effects in terms of possible void defect formation at the Be/DT interface at the leakage sites, even for room temperature measured half-lives of greater than 7 days, and even up to a year or more, which is beyond the current leakage measurement capability. These may, however, be ameliorated by void diffusion in the solid.

Based on the estimates presented here, the current leak rate specification for Be shells may lead to capsule leakage which negatively impacts ignition experiments. Development of techniques to produce leak-free Be shells as well as techniques to determine leakage time constants in excess of a year is clearly desirable. In addition, confirmation of the existence of voids in the solid DT, or determining the reasons they do not form, is needed to provide confidence that ignition experiments will be successful.

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